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MAGNESIUM—LITHIUM ALLOYS

A REVIEW OF CURRENT DEVELOPMENTS

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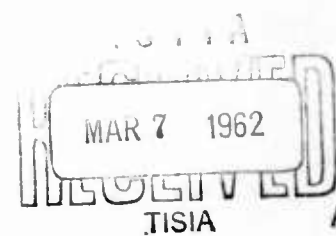
MAGNESIUM-LITHIUM ALLOYS
A REVIEW OF CURRENT DEVELOPMENTS

P. D. Frost*

INTRODUCTION

Magnesium-lithium-base alloys are of interest for several reasons: First, by virtue of the lithium, which has a specific gravity of 0.53, they have lower densities than any commercial magnesium alloy. Second, lithium markedly improves the ductility and workability of magnesium. Third, since they have approximately the same modulus of elasticity as magnesium alloys, namely about 6.5 million psi, the magnesium-lithium alloys have a high ratio of elastic modulus to weight, making possible rigid, light structures.

Research has been in progress on structural magnesium-lithium alloys since about 1944 at Battelle, The Dow Chemical Company, Armour Research Foundation, Case Institute, Frankford Arsenal, and abroad in England and in Russia. Prior to 1944, research on phase diagrams had been conducted in Germany and in Russia. As yet, no magnesium-lithium alloys are in commercial production. However, their unique properties keep these alloys of interest for future applications.

The purpose of this memorandum is to review very briefly the progress that has been made in the development of these alloys.

PHASE RELATIONSHIPS

Magnesium-lithium phase relationships have been studied by many investigators. The most widely used diagram has been that of Grube, shown in Figure 1[1]**. However, in 1954, Freeth and Raynor redetermined the diagram as shown in Figure 2[2].

The binary system is characterized by two phases. The magnesium solid solution, having a hexagonal close-packed structure, exists from 0 up to about 5.7 weight per cent (17 atomic per cent) of lithium. Between 5 and 11 weight per cent, a two-phase region exists which comprises hexagonal close-packed and body-centered cubic solid solutions. Beyond about 11 per cent (30 atomic per cent) lithium, the lithium solid solution, which is entirely body-centered cubic, is stable. At the present time the body-centered cubic alloys containing up to approximately 15 per cent lithium are of greatest research interest because of their extreme lightness and excellent ductility. In these materials the good ductility results from the inherently ductile lattice structure of the body-centered cubic system as compared with that of the hexagonal close-packed system.

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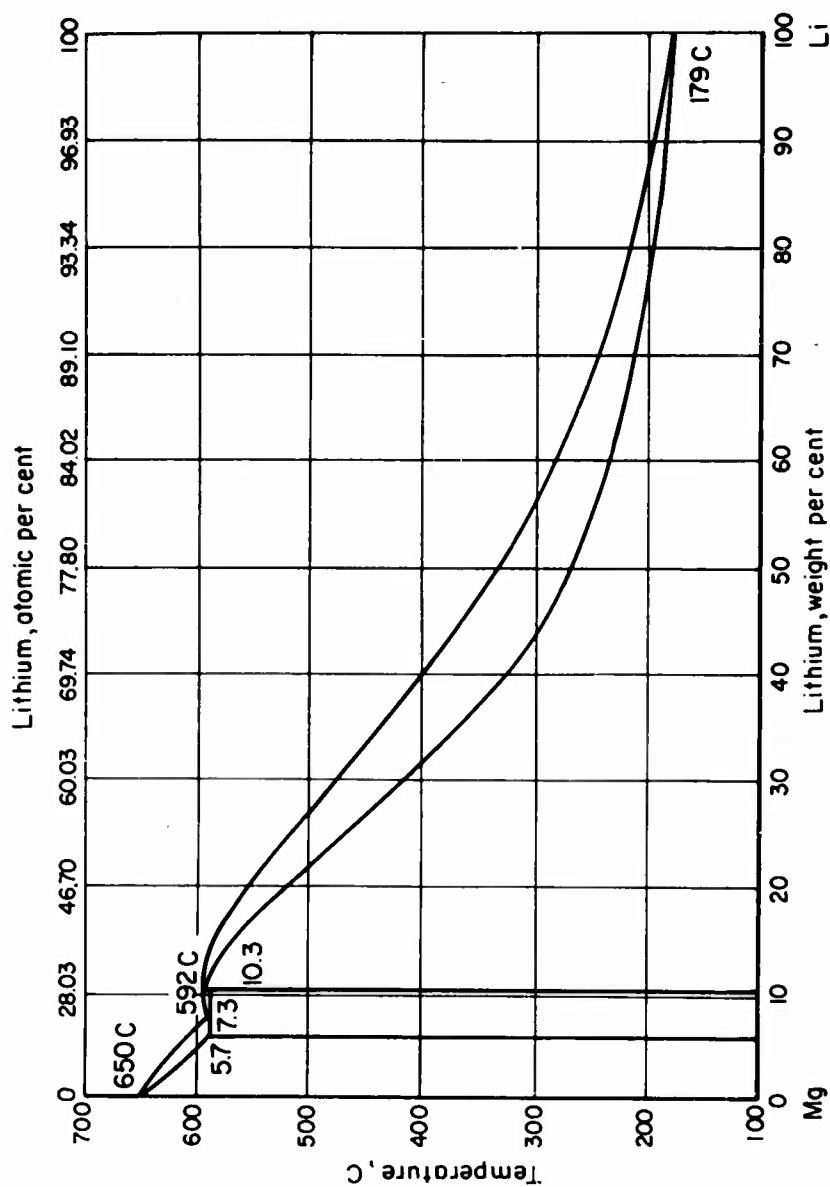
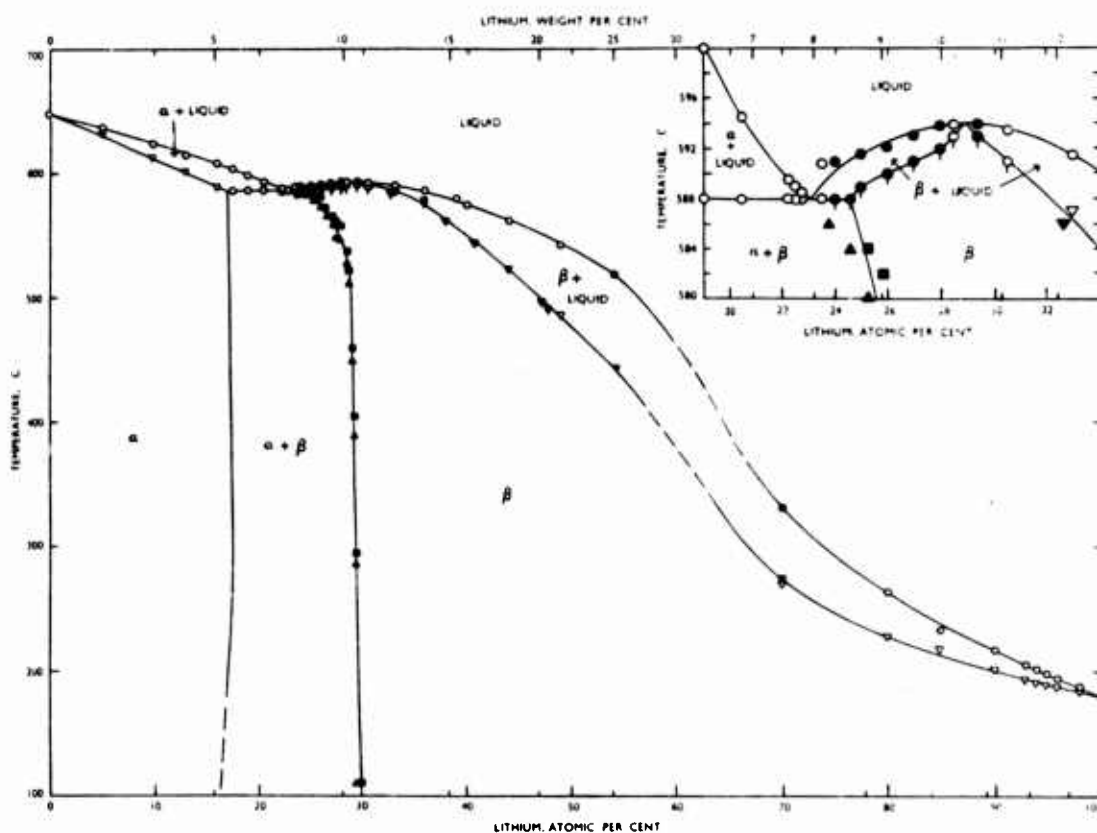FIGURE 1. MAGNESIUM–LITHIUM EQUILIBRIUM DIAGRAM (GRUBE)

FIGURE 2.   MAGNESIUM–LITHIUM PHASE DIAGRAM ACCORDING
TO FREETH AND RAYNOR

In this regard lithium also increases the ductility of the hexagonal magnesium solid solution. It has been known for many years that lithium is the only element capable of decreasing the c/a ratio of the magnesium lattice. The effects of lithium and other elements on the magnesium lattice were studied quantitatively by R. S. Busk[3]. Very recently, Hauser, et al., showed that the presence of relatively small amounts of lithium in the hexagonal lattice of magnesium permit the lattice to deform by slip in the prismatic planes as well as in the basal planes.[4] Hauser related this effect to the capability of lithium to reduce the c/a ratio of the lattice. Of course, when enough lithium is present to convert the lattice from hexagonal to the body-centered cubic structure, the extreme ductility resulting therefrom is derived from the many slip systems possible in the body-centered cubic lattice.

The principal metallographic phases of magnesium-lithium binary alloys are shown in Figure 3.

BASIS FOR TERNARY AND COMPLEX ALLOYS

Magnesium-lithium binary alloys, although light and ductile, are not strong. The greatest part of the research effort carried out between 1944 and the present time has been to develop high-strength alloys. Several complex systems of magnesium-lithium-base alloys are strengthened by precipitation hardening. The alloying elements of greatest interest have been aluminum, zinc, cadmium, and silver. These elements have shown the greatest solubility in the magnesium-lithium matrix and also the greatest ability to strengthen the matrix by precipitation hardening.

In this connection it is interesting to consider these elements in relation to others of the periodic table as they, in turn, are related to lithium. Hume-Rothery[5] and others have observed that the ability of one element to dissolve in the solid lattice of another element is governed by several factors. These include the type of crystal structure, the relative size of the dissolved atom with respect to the solvent atom, the valences of the metals, and the relative electronegativities of the metals. For complete mutual solubility, the metals must have the same crystal structure. However, if their atomic radii do not differ by more than about 15 per cent they can have different crystal structures and still have appreciable solid solubility. It is also known that elements which have electronegativity values that do not differ by more than approximately ±0.4 units tend to have greater mutual solubility.[6] In Figure 4 an attempt has been made to show the relationships between solubility and the factors of atomic radii and electronegativity as these factors apply to the lithium atom. This plot must be considered strictly as a guide for the development of alloys and not quantitatively. The lithium atom is shown at the center of the plot with the 15 per cent size-factor limits favorable to extensive solubility in the lithium lattice. The electronegativities of the various elements are plotted in relationship to that of lithium. Most elements are, of course, more electronegative than lithium and most are outside the ±0.4 limits. However, on the basis of size factor alone, many metals might be expected to dissolve in the lithium lattice.
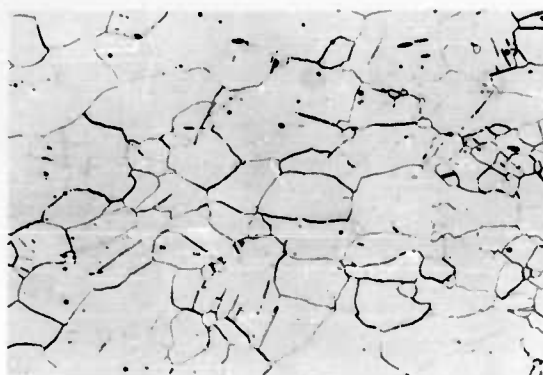
Alloy A. Mg-3% Li
 Specific gravity, 1.66
 Microstructure, α phase HCP
 Tensile strength, 20,200 psi
 Yield strength, 12,500 psi
 Elongation, 23 per cent
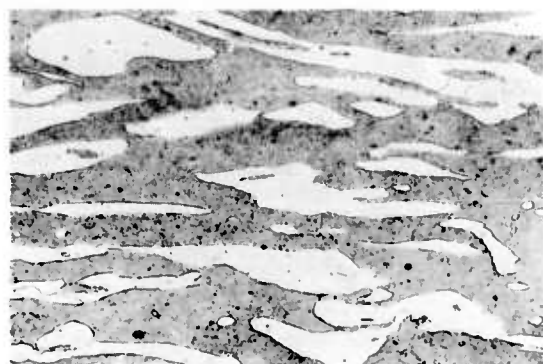
Alloy B. Mg-9.1% Li
 Specific gravity, 1.51
 Crystal structure, α (HCP) and
  β (BCC)
 Tensile strength, 18,300 psi
 Yield strength, 13,900 psi
 Elongation, 36 per cent
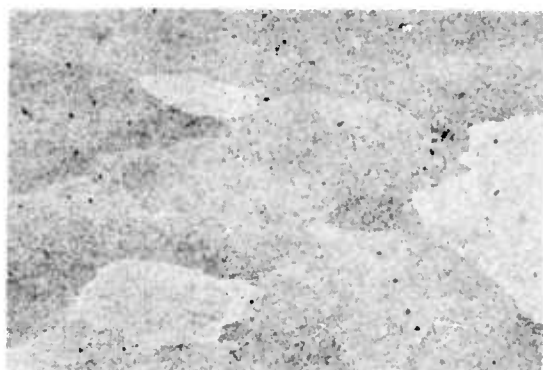
Alloy C. Mg-16.7% Li
 Specific gravity, 1.33
 Crystal structure, β phase BCC
 Tensile strength, 14,500 psi
 Yield strength, 10,300 psi
 Elongation, 34 per cent

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FIGURE 3.   EFFECTS OF LITHIUM IN MAGNESIUM-LITHIUM BINARY ALLOYS

Note: The specific gravity of commercial unalloyed
magnesium is about 1.74. Rolled, annealed sheet,
comparable to the form of these alloys, has a
tensile strength of about 27,000 psi with elongation
of about 15 per cent.
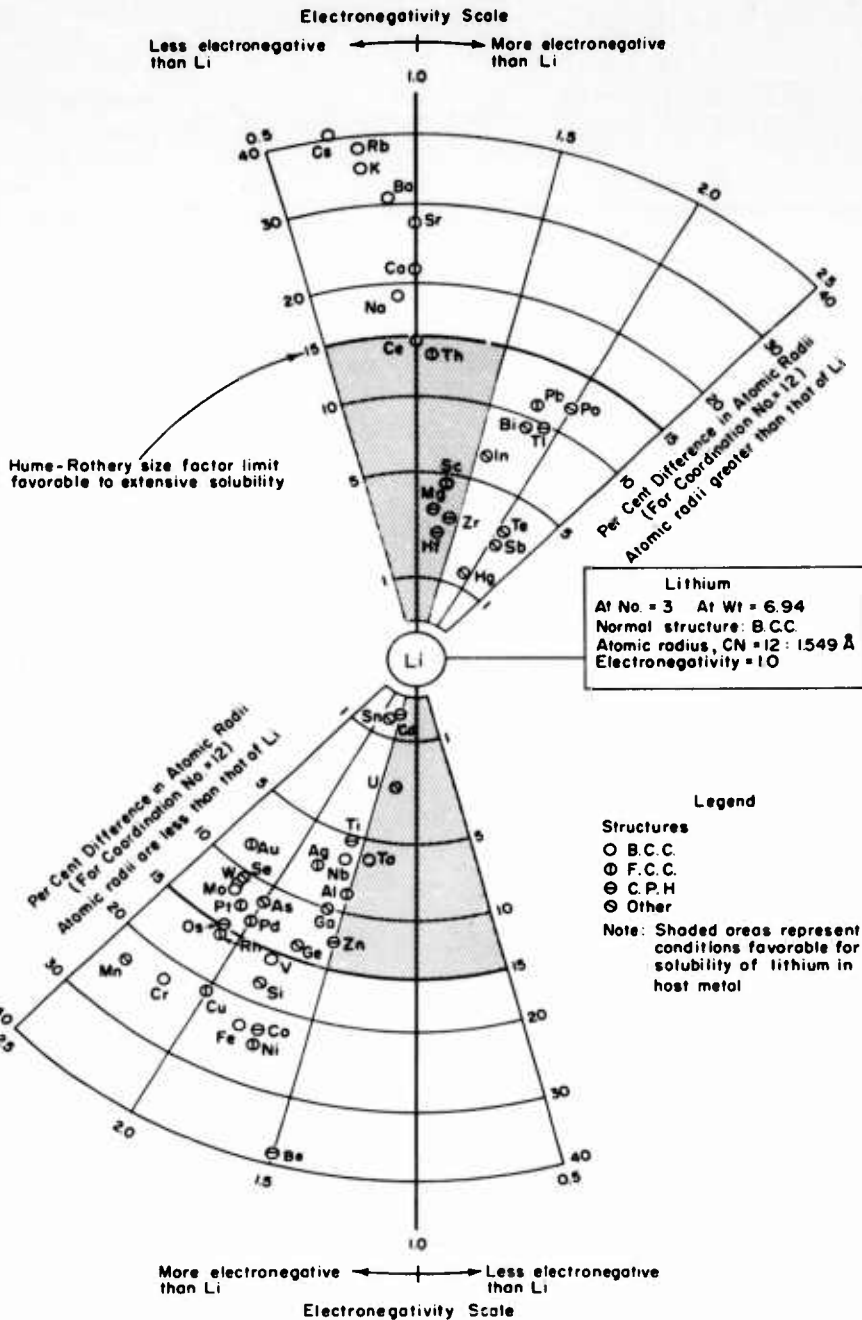
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FIGURE 4. SCHEMATIC DIAGRAM SHOWING DIFFERENCES BETWEEN
ATOMIC RADII AND ELECTRONEGATIVITIES OF LITHIUM
AND OTHER ELEMENTS AND THEIR EFFECTS ON SOLUBILITY

Per cent difference in atomic radii is calculated
for lithium as solute.

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The shaded area of Figure 4 represents those elements which have both favorable atomic size and electronegativity. It is seen that magnesium falls well within the shaded zone. Cadmium, zinc, and aluminum, likewise, are within or close to this shaded zone. Silver falls outside of the shaded zone, but its position can be shifted into the zone depending on the valence assumed for this metal. It might be added that mercury, indium, and thallium, whose atomic diameters are favorable for solubility, all exhibit appreciable solubility in the magnesium-lithium-base solid solution.

Figure 5 shows the effects of zinc on the hardness of a magnesium-lithium alloy containing approximately 14 per cent lithium.[7] Also shown in the figure is the effect of prolonged aging at 200 F on the hardness of the magnesium-lithium-zinc alloys. All alloys were solution treated 1 hour at 700 F and quenched rapidly in toluene at minus 105 F. It will be seen that the binary alloy has very low hardness but that the addition of 2 per cent zinc produces a very substantial increase in the hardness. This amount of zinc does not cause any age-hardening reaction at 200 F, the hardening mechanism being strictly that of strengthening the solid solution. With the addition of from 4 to 10 per cent zinc, the hardness (and strength) of the alloys is progressively increased in the as-quenched condition. At the same time further hardening is achieved by the aging treatment. This precipitation hardening is caused by the formation of a transition phase (theta) discovered a number of years ago at Battelle. The existence of this phase has been confirmed by investigators at Dow Chemical Company and elsewhere.[8,9] It has the composition $MgLi_2X$ where X may be zinc, aluminum, cadmium, or silver, and possibly other elements. With amounts of zinc larger than about 10 per cent, the hardening mechanism cannot be supressed by the rapid quench, and the hardness of the quenched alloys exceeds 100 Rockwell E. These alloys harden still further with an aging treatment. In the as-quenched condition or at the peak hardness, the alloys containing over 12 per cent zinc are quite strong and brittle. In this type of alloy the tensile strength has been known to reach 70,000 psi with compressive strengths even higher. However, in this condition the alloys have practically no ductility. With prolonged aging at 200 F, or even as low as 150 F, the alloys gradually lose their strength. When the zinc content is not too high, they recover much of their original ductility after such an aging treatment.

The problem that has beset the magnesium-lithium alloys ever since the first research, has been to prevent this overaging or softening effect at relatively low temperatures (from room temperature to 200 F). It was found at Battelle and confirmed by other investigators that the instability of mechanical properties is associated with the transitory nature of the theta phase. Dow found that this phase disappears on heating to relatively low solution temperatures.

Many combinations of alloying elements have been investigated and the work has been carried out by a number of researchers. Certain elements, especially silver, extend the hardness curves so that aging does not occur at 150 F as rapidly as with zinc or aluminum. An example of the aging behavior of one alloy containing approximately 12 per cent lithium, 15 per cent cadmium, and 5 per cent silver is shown in Figure 6.[8] It will be observed that this alloy can be solution treated and quenched to relatively low hardness levels, namely 70 Rockwell E. Aging at 150 F produces a typical age-hardening curve
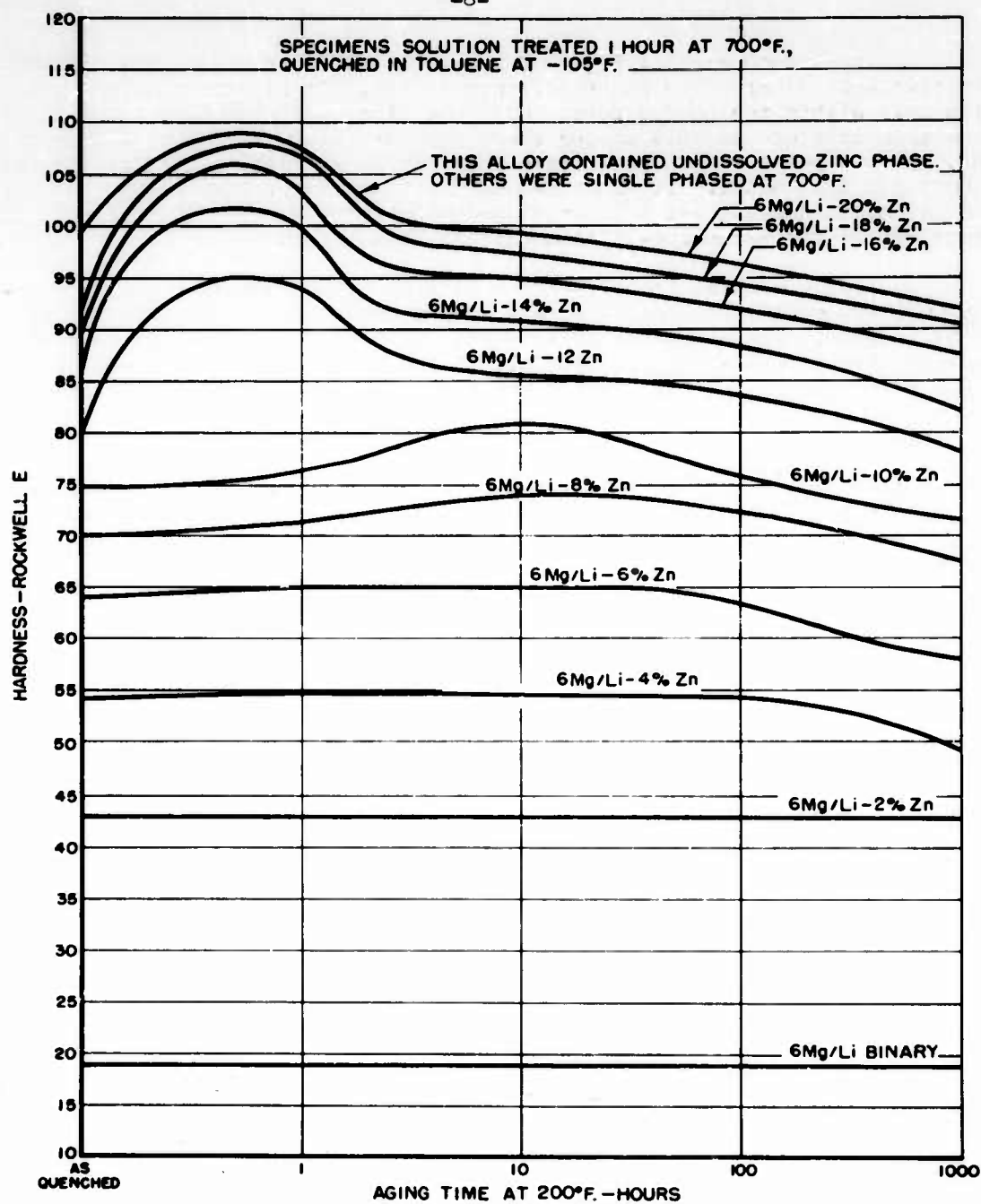
FIGURE 5. EFFECT OF ZINC CONTENT AND AGING TIME AT 200 F
ON HARDNESS LEVEL OF Mg-Li-Zn ALLOYS HAVING THE
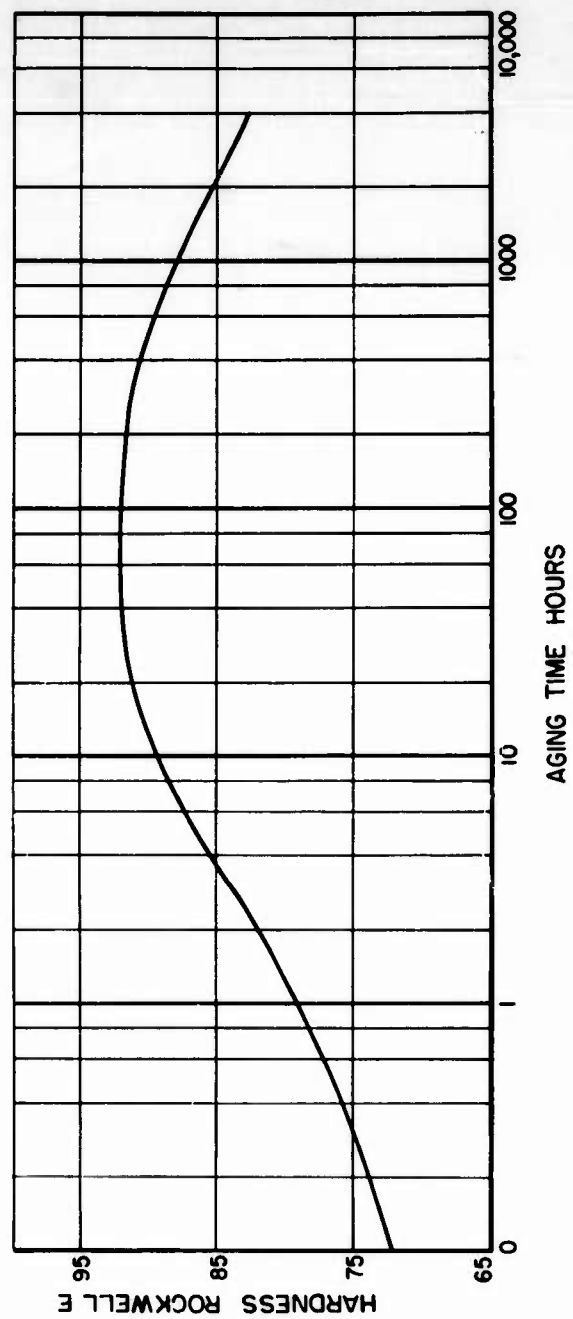SAME RATIO OF MAGNESIUM TO LITHIUM CONTENTS[6]

FIGURE 6. AGE-HARDENING BEHAVIOR OF A Mg-12%Li-15%Cd-5%Ag ALLOY AT 150 F FOLLOWING A SOLUTION TREATMENT AT 500 F(7)

with the maximum hardness being reached in about 20 hours. In this regard a hardness of about 85 Rockwell E is representative of ultimate strength levels of the order of 45 to 50,000 psi, yield strengths of approximately 35 to 40,000 psi, and elongations of 10 to 15 per cent. Thus, this alloy is capable of being heat treated to develop excellent formability as quenched, and excellent mechanical properties as aged. Nevertheless, it can be seen that continued aging at 150 F produces a decrease in strength.

In regard to the alloys containing silver, reference is made to recent work conducted by Jones and Hogg[10] at Cardiff University. These authors reported on an extensive research study on the effects of various alloying elements on the properties and stability of the magnesium-lithium-base alloys. They observed that silver was most effective in increasing stability, and concluded that instability is related to the valency of the alloying elements or the electron to atom ratio of the alloy. Silver, added to decrease the electron concentration, produced the greatest stabilizing effects of the alloys studied. This work is of more recent origin than the extensive research conducted at Battelle and Dow. However, although it has contributed an interesting hypothesis to the general knowledge, the authors point out that the work did not solve the instability problem.

ALLOYS OF GREATEST CURRENT INTEREST

Although no one has yet been successful in developing a stable, high-strength magnesium-lithium alloy, research in the meantime has been directed toward applications for lower strength, more dilute alloys, which retain their room-temperature properties for indefinite periods of time. Such alloys either do not age harden extensively or they can be overaged or stabilized by a thermal treatment. They do not contain sufficient quantities of a third element to render them brittle in the as-quenched condition and are, therefore, comparatively very ductile. The most interesting system is the magnesium-lithium-aluminum system. Figure 7 shows the aging character-istics, following a solution heat treatment, of a series of ternary magnesium-lithium-aluminum alloys.[7] Hardness is plotted as a function of aging time at 200 F. As in the magnesium-lithium-zinc system, the alloys containing less than about 1 per cent aluminum do not respond to the aging cycle at 200 F (aging at room temperature would produce some response in the alloy containing 0.5 per cent aluminum). Those alloys which contain more than about 2 per cent aluminum are comparatively hard and brittle as quenched. They overage in a relatively few hours to a state of reasonable ductility. Depending on their condition of aging, the alloys can have strengths ranging from 20,000 psi up to 70,000 psi with ductilities in inverse proportion to their strength. The exceedingly high strength condition develops a very brittle material.

Because we have not learned to stabilize the higher strength alloys, the magnesium-lithium-aluminum alloys containing approximately 1 to 2 per cent aluminum and about 14 per cent lithium, and which have moderate strength, are of greatest interest for possible applications in the future. During the last two years the George C. Marshall Space Flight Center (formerly
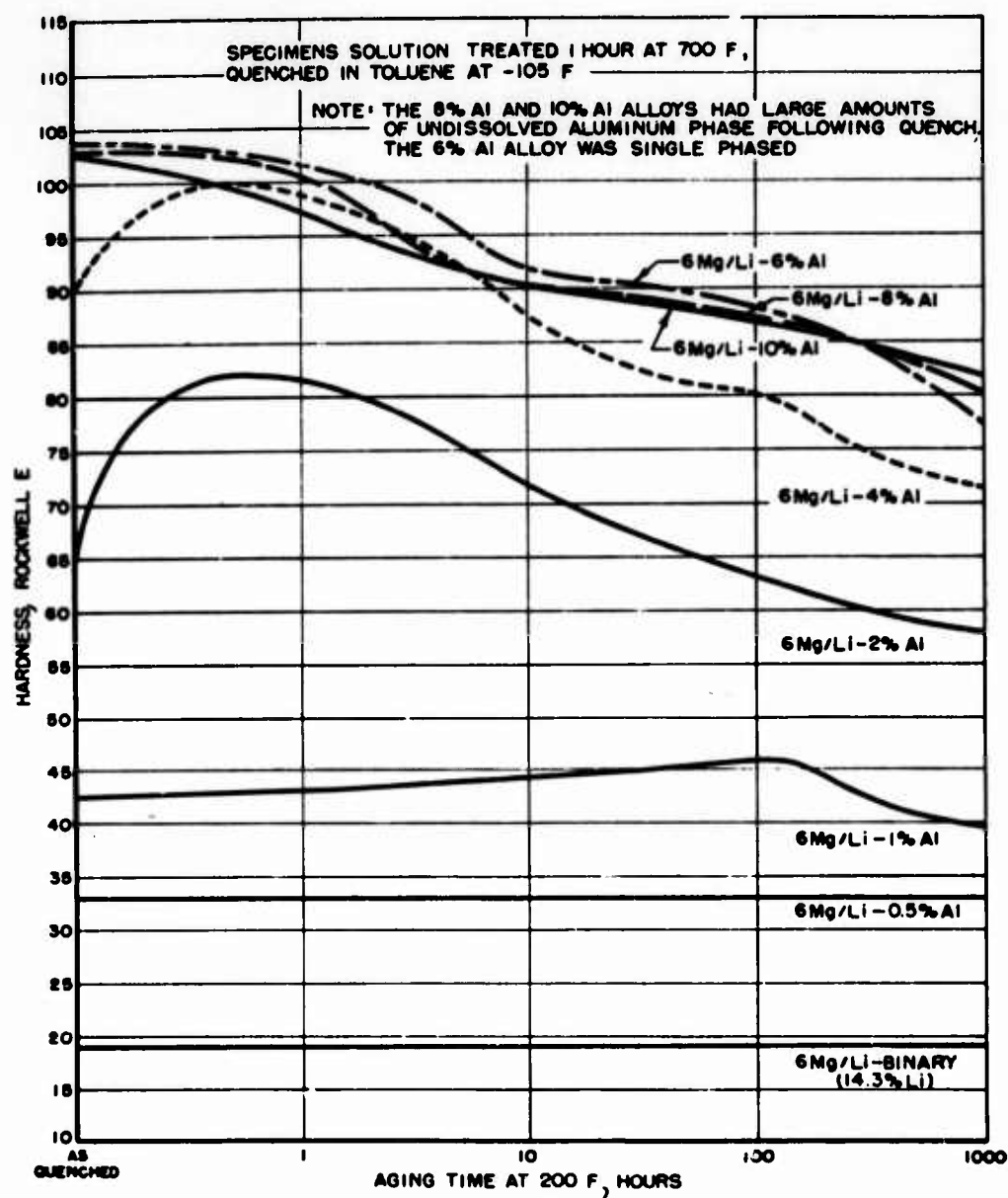
FIGURE 7. EFFECT OF ALUMINUM CONTENT AND AGING TIME AT
200 F ON HARDNESS LEVEL OF Mg-Li-Al ALLOYS
HAVING THE SAME RATIOS OF MAGNESIUM-TO-LITHIUM
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the Army Ballistic Missile Agency) has sponsored a study to evaluate magnesium-lithium alloys for potential usage in missiles, satellites, and space vehicles.[11] During this research the mechanical properties of various alloys were determined. Rather extensive property evaluation work was conducted on two alloys having moderate strength: LA141, containing about 14 per cent lithium and 1 to 2 per cent aluminum, and LA91, containing 9 per cent lithium and 1 to 2 per cent aluminum. The first alloy is body-centered cubic in structure while the latter alloy has a mixed body-centered cubic and hexagonal close-packed structure.

The alloys were melted under a LiCl-LiF flux according to previously published practice[8], and properties were determined on forged and rolled sheet and on extrusions. A brief summary of some of the mechanical and physical properties of the alloys are reported in Tables 1 and 2.

The alloys were cold rolled to ascertain the effects of cold work on the mechanical properties. A maximum strength of the order of 33,000 psi with a yield strength of 30,000 psi and elongations of around 16 per cent are obtained by cold working.

One part of this research was an exploratory program to study improved methods for melting the alloys. Flux melting of heats weighing much more than 100 pounds is cumbersome. In the past, the alloys have sometimes been melted in closed iron crucibles under an argon atmosphere. For the NASA investigation, techniques were worked out for melting 100-pound heats of the alloys under a reduced pressure of argon, namely about 50 millimeters Hg. Melting was carried out in a vacuum melting furnace which has a capacity of 300-pound heats of steel. Both the iron crucible and the mold were contained in the vacuum system during melting and pouring. The furnace was evacuated to 10 microns pressure or less and back filled with argon to the desired pressure. The mold was preheated, the charge melted, and ingots weighing up to about 90 pounds were poured without difficulty. After cropping, the ingots had dimensions of approximately 10 x 10 x 12 inches of sound, gas-free metal. While this method is satisfactory for heats of this size, it would not be economical for production quantities without major modifications.

As part of the NASA contract, an exercise in fabrication of the magnesium-lithium-aluminum alloy sheet was conducted by welding a prototype vessel of the type that might be similar in size to a satellite. For this work, sheet approximately 0.090 inch thick was required for a cylindrical center section 30 inches in diameter by 45 inches long. The ends of the cylinder were to be capped with dish-shaped bulkheads approximately 6-3/4 inches deep.

The ingots described earlier were homogenized 48 hours at 550 F and forged on a 700-ton vertical hydraulic press. Forging was carried out at 550 F. The ingots were upset to rolling slabs approximately 12 x 18 x 5 inches. These were machined and hot rolled at 550 F on a 16 x 24-inch, two-high, reversible experimental mill to plate 1/4 inch thick. This material was finished-rolled to sheet, and the end sections of the vessel were formed in a commercial shop. Plate sections 1/4 x 18 x 48 inch were cross rolled at 450 F to sheet 0.090 inch thick by 48 inches square for the bulkheads. These were flattened under heavy weights at 250 F, cleaned in an alkali bath and in chromic acid, and then coated with a thin film of graphite prior to forming. The bulkhead sheets were formed at 400 to 500 F. Figure 8 shows the press and one of the formed parts.

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TABLE 1. EFFECT OF FABRICATION VARIABLES ON TENSILE PROPERTIES OF AS-FABRICATED LA141 AND LA91 ALLOYS

Method of Fabrication	Fabrication Procedure	LA141					LA91				
		Ultimate Tensile Strength, psi	Yield Tensile Strength, psi	Elongation(a), per cent	Reduction in Area, per cent	Hardness(b), R_E	Ultimate Tensile Strength, psi	Yield Tensile Strength, psi	Elongation(a), per cent	Reduction in Area, per cent	Hardness(b), R_E
As extruded	0.187 x 2 x 2-inch T-section extruded from 4-7/16-inch-diameter billet at 500 F	30,200	28,700	11	--	80	20,900	18,100	31	--	61
		29,600	28,200	10	--		21,100	18,100	30	--	
As extruded and rolled	0.187 x 2-inch flange of T-section rolled at 450 F to 0.063-inch-thick sheet	32,500	30,400	19	--	78	28,600	24,000	18	--	70
		32,400		15	--		27,600		18	--	
As forged	Bar stock hammer forged at 550 F from cast ingot	31,500	3),000	14	36	--	23,800	20,800	39	62	--
		31,700		10	17		23,300		39	62	
As forged and rolled	Forged bar rolled at 450 F to 0.063-inch-thick sheet	25,900	24.00	18	--	67	30,000	20,200	34	--	72
		26,200		18	--		29,000		34	--	
As rolled	As-cast ingot rolled at 450 F to 0.063-inch-thick sheet	28,100	26,400	19	--	65	31,100	24,900	15	--	72
		28,300		16	--		30,600		15	--	
As rolled	As-cast ingot rolled at 450 F to 0.090-inch-thick sheet, followed by cold rolling to 0.063-inch-thick sheet (30 per cent cold reduction)	31,300	29,500	18	--	71	33,400	26,800	20	--	78
		31,800		18	--		34,000		20	--	

(a) Tensile specimens:

As extruded – Standard plate specimen 0.187 inch thick; reduced section 1-1/4 inches wide by 8 inches long.

As forged – Substandard round specimen 7/16 inch in diameter; reduced section 1/4 inch in diameter by 1 inch long.

As rolled – Standard sheet specimen 0.063 inch thick; reduced section 1/2 inch wide by 2 inches long.

(b) Values are averages of three readings.

TABLE 2. TYPICAL PROPERTIES OF MAGNESIUM–LITHIUM ALLOYS

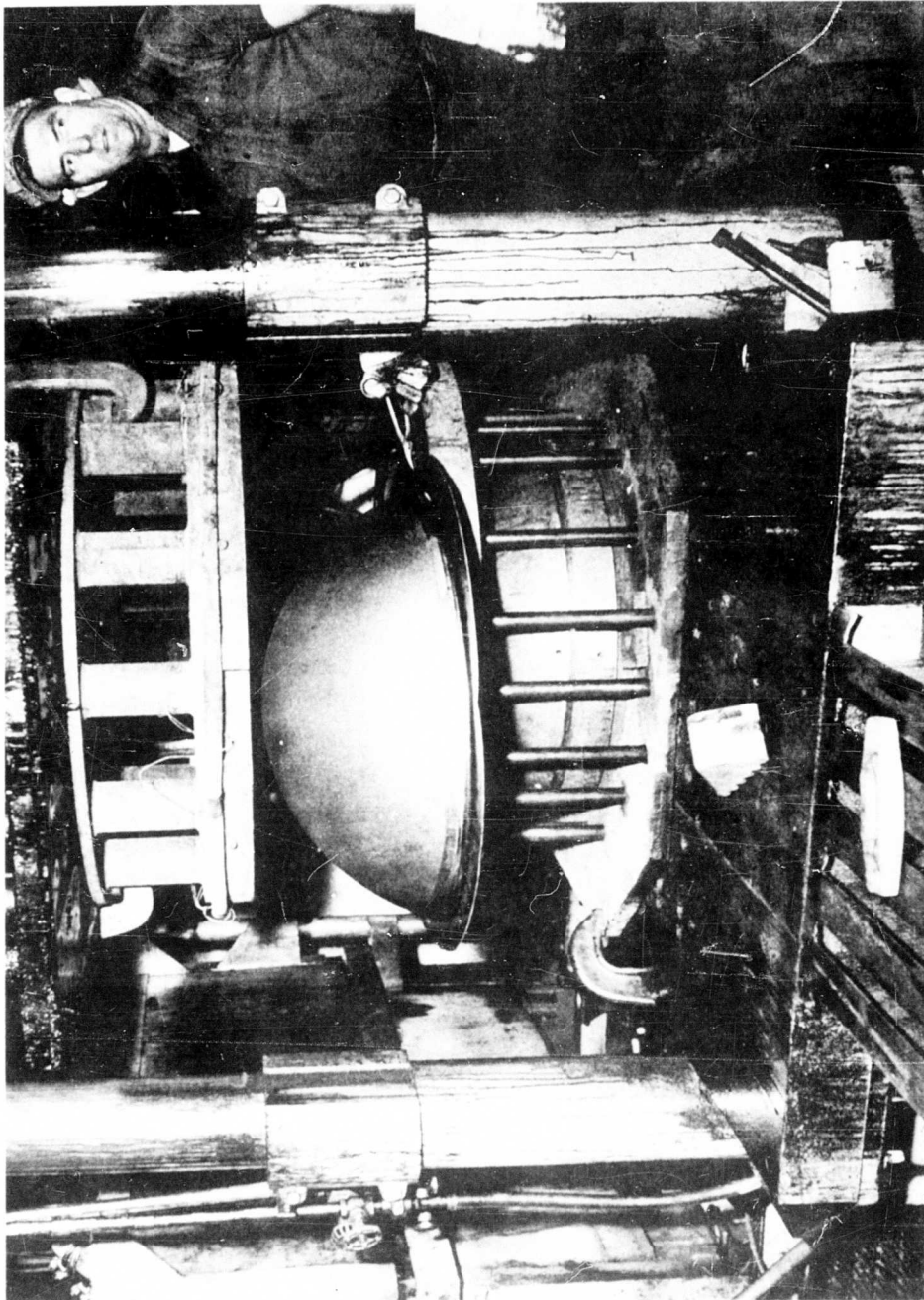|	LA141	LA91
Nominal Composition, weight per cent	Mg–14.1Li–1.5Al	Mg–9Li–1Al
Type of Structure	Beta (bcc)	Alpha + beta (hcp + bcc)
Density, g/cm^3	1.35	1.45
Coefficient of Mean Linear Thermal Expansion (68–200 F), 10^{-6} in./in./F	21	18
Specific Heat (32–300 F), Btu/lb/F	0.34 – 0.36	0.28 – 0.33
Thermal Conductivity (50–300 F), $Btu/ft/hr/ft^2/F$	--	32.3
Normal Spectral Emissivity of Vapor-Blasted Sheet at 360 K		
4–Micron Wavelength	0.55	0.45
13–Micron Wavelength	0.50	0.37
Rate of Weight Change in Vacuum at 800 F, $\mu g/cm^2/sec$	3.8×10^{-2} at 2×10^{-6} mm Hg	1.1×10^{-2} at 1×10^{-6} mm Hg
Corrosion Rate, 60–Day Exposure to 100 Per Cent Humidity at 95 F		
Uncoated	10 mdd[a]	9 mdd
Anodized	1.6 mdd	1.7 mdd
Mechanical Properties		
Ultimate Strength, pdi	21,000	22,000
Yield Strength, psi	17,000	16,500
Elongation, per cent in 2 in.	25 – 30	30 – 35
Bend Radius	1T	1T
Hardness, R_E	65	55
Modulus of Elasticity, 10^6 psi	6.5	6.6

(a) mdd = $mg/dm^2/day$.

FIGURE 8. DRAWN 32-INCH-DIAMETER BULKHEAD SECTION. LA141 ALLOY
Sheet is 0.090 inch thick; courtesy Brooks and Perkins, Inc.

The vessel was assembled by welded construction at NASA. Welding was done by the MIG process using EZ-33A alloy filler wire (Mg-2.7Zn-0.7Zr-3 Rare Earths). Welds were made with a straight closed-butt joint design at a speed of 40 inches per minute. These were single-pass welds with the exception of areas where manual repairs were necessary. The finished vessel is shown in Figure 9. The mechanical properties of weld-test coupons of both LA141 and LA91 alloys as welded were about 20,000 psi ultimate strength, 15,000 psi yield strength, and 7 per cent elongation. Actually the ductility of welds in these alloys can be doubled by a thermal stabilizing treatment after welding. The bend ductility of stabilized welds in sheet is 1t.

OTHER APPLICATIONS

In addition to possible usage in space, the magnesium-lithium alloys have shown great promise as an armor material for defeating shell fragments (in this regard the alloys might also be considered for protecting space vehicles from micrometeorites). Research has been carried on for a number of years on light aircraft armor. It has been found that the magnesium-lithium alloys containing around 13 to 14 per cent lithium plus 1 to about 6 per cent aluminum have excellent armor characteristics. On a weight basis they afford better protection against low- and intermediate-velocity fragments than any other metallic material studied to date.[12]

During the last several years the Detroit Tank Arsenal, collaborating with Frankford Arsenal and the Dow Metal Products Company, has adopted and modified these alloys to evaluate tham as a material for the body of a large armored vehicle. This vehicle, the M113, is a personnel carrier having a capacity of 12 men plus the driver. It is air droppable and must therefore be as light as possible. The development work carried out at the Dow Metal Products Company has involved melting ingots weighing over 1000 pounds and rolling them to plate up to 3 inches thick. It has also necessitated welding thick plates and protecting them against corrosion. Details of the Dow work are not published. However, it was found that it was not practical to attempt to weld alloys containing as much as 6 per cent aluminum. Therefore, the aluminum level of 1 to 2 per cent was adopted. The optimum composition is approximately 13.5 per cent lithium, 1 to 2 per cent aluminum, 0.1 per cent manganese (for corrosion resistance), and 0.005 per cent iron, maximum. Welding is best accomplished by the use of a commercial magnesium alloy filler material such as AZ92 (Mg-9Al-2Zn). Welds must be stabilized by a thermal overaging treatment for proper ductility.

Corrosion protection has been accomplished by the application of the new stannate treatment, details of which are available from Dow. Figure 10 is a photograph of the vehicle body.

The M113 vehicle is at the present time the most advanced development for the magnesium-lithium alloys. Numerous inquiries are being made concerning the recent developments in the alloys, and it is known that several organizations involved in space-flight development have the alloys under study.
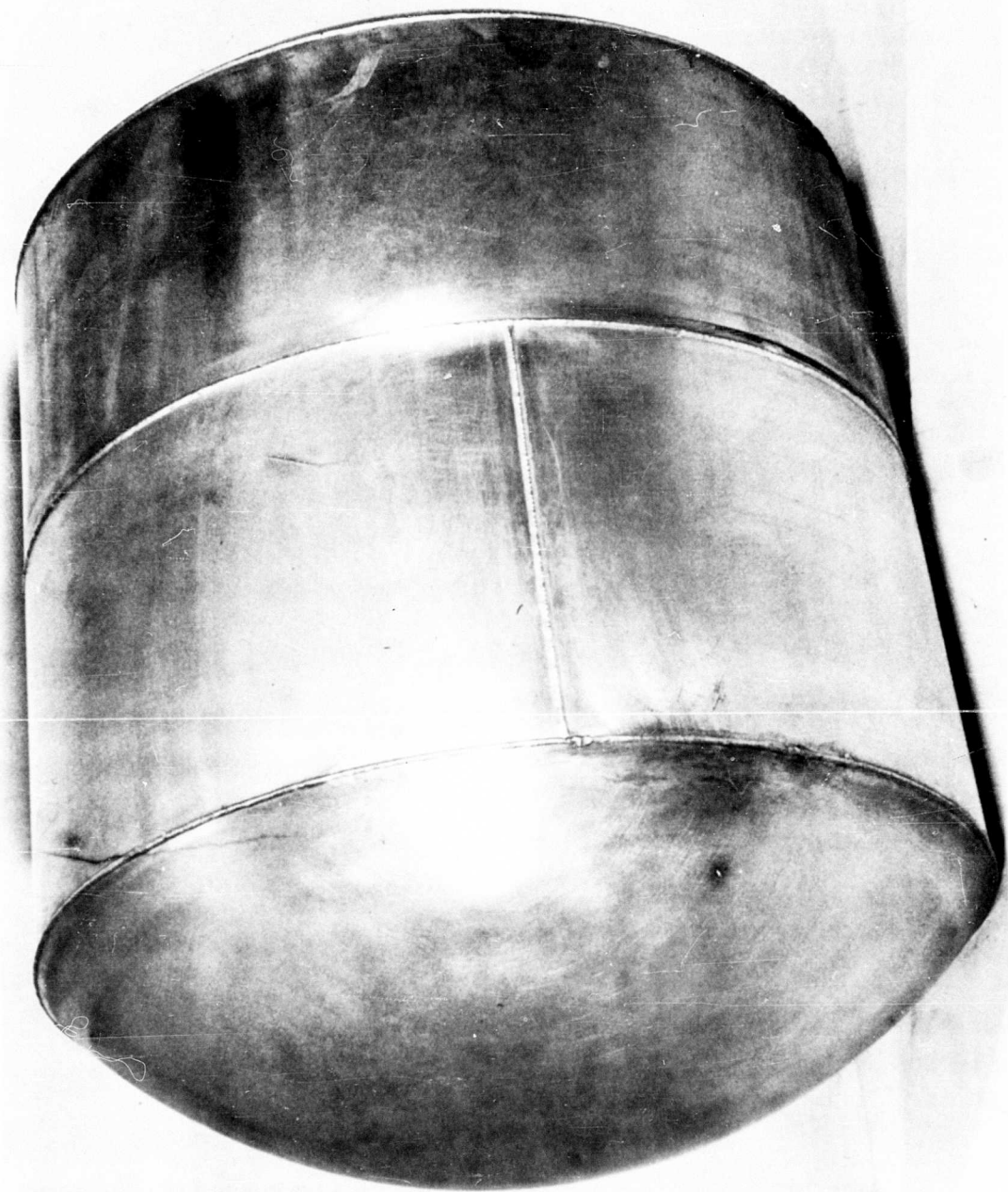
FIGURE 9. VESSEL FABRICATED AT NASA FROM LA141 AND LA91 ALLOYS

Cylindrical section measures 30 inches in diameter and 30 inches deep. Photograph courtesy of NASA.
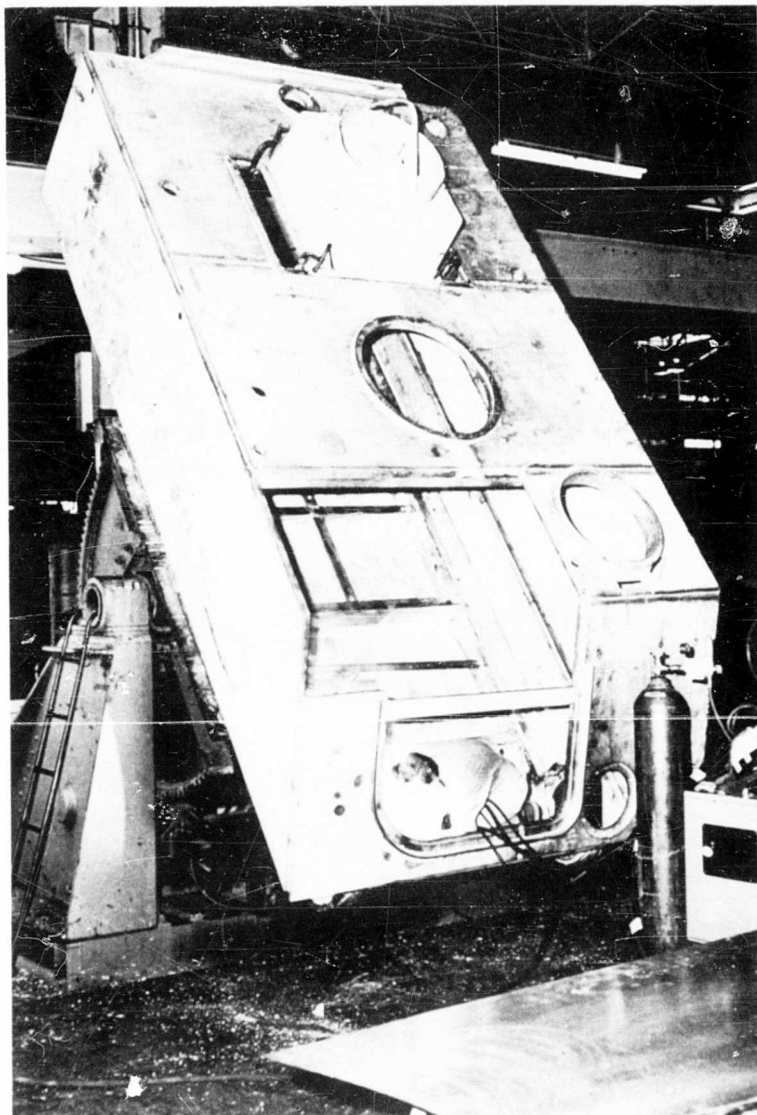
FIGURE 10.   PROTOTYPE BODY OF THE M113 ARMORED VEHICLE
MADE FROM A MAGNESIUM-LITHIUM ALLOY

Courtesy Detroit Tank Arsenal

Most likely the first applications for the alloys will involve parts which
do not require high structural strength, such as instrument cases, enclosures
such as the prototype vessel built at NASA, and shields for protection against
micrometeorites and shell fragments. It is expected that these specialized
military and space applications will precede any commercial uses of the alloys.

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119	The Emittance of Iron, Nickel, Cobalt and Their Alloys, July 25, 1961, (AD 261336 $2.25)
120	Review of Recent Developments on Oxidation-Resistant Coatings for Refractory Metals, July 31, 1961, (AD 261293 $0.50)
121	Fabricating and Machining Practices for the All-Beta Titanium Alloy, August 3, 1961, (AD 262496 $0.50)
122	Review of Recent Developments in the Technology of Nickel-Base and Cobalt-Base Alloys, August 4, 1961, (AD 261292 $0.50)
123	Review of Recent Developments in the Technology of Beryllium, August 18, 1961, (AD 262497 $0.50)
124	Investigation of Delayed-Cracking Phenomenon in Hydrogenated Unalloyed Titanium, August 30, 1961
125	Review of Recent Developments in Metals Joining, September 1, 1961, (AD 262905 $0.50)
126	A Review of Recent Developments in Titanium and Titanium Alloy Technology, September 15, 1961
127	Review of Recent Developments in the Technology of Tungsten, September 22, 1961
128	Review of Recent Developments in the Evaluation of Special Metal Properties, September 27, 1961
129	Review of Recent Developments in the Technology of Molybdenum and Molybdenum-Base Alloys, October 6, 1961
130	Review of Recent Developments in the Technology of Columbium and Tantalum, October 10, 1961
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136	Fabrication of Tungsten for Solid-Propellant Rocket Nozzles, November 2, 1961
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144	Review of Recent Developments in Titanium Alloy Technology, December 29, 1961
145	The Use of Nickel-Base Alloys in the Rotating Parts of Gas Turbines for Aerospace Applications, January 11, 1962